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INTENSITY DEPENDENCE OF THE THIRD HARMONIC GENERATION EFFICIENCY FOR HIGH POWER FAR INFRARED RADIATION IN N-SILICON

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Abstract:

The third harmonic generation of far-infrared (FIR) laser radiation in n-doped Silicon was measured for the first time with full temporal resolution of the power fluctuations during the FIR laser pulse. Thus the intensity dependence of the nonlinear coefficient $\chi^{(3)}$ could be observed. For 2MW incident power, corresponding to an intensity of 15MW/cm^2, at a wavelength of 676μm a power conversion efficiency of 10^{-3} was reached.

Introduction:

In the far-infrared (FIR) regime the linear as well as the nonlinear optical properties of doped semiconductors are mainly determined by the motion of the free carriers (i.e. electrons in the conduction band for n-doped materials) caused by the electric field of the incident wave. The measurement of the nonlinear optical properties in the FIR thus provides the possibility to investigate nonlinearities of the free electron system. The FIR frequencies are well below the absorption bands in the mid infrared regime which are due to optical phonons. First experiments on the frequency tripling of FIR-radiation in semiconductors were reported by Mayer et al. [1]. However they were limited by their laser power and their detection system, which allowed only time-integrated measurements of the frequency tripling efficiency, averaged over the laser pulse. Because of transversal mode beating the pulses of high power FIR lasers usually show strong temporal fluctuations in power. This makes the interpretation of time-integrated measurements difficult if the investigated process depends nonlinearly on the incident power. The time-resolved frequency tripling experiments described here overcome this problem by directly measuring the instantaneous power at both the fundamental and the third harmonic frequencies.

Our experiments are of practical interest for the development of an efficient frequency converter for high power FIR radiation. In contrast to the optical and near infrared frequencies where frequency converters became standard tools in many applications, the FIR regime was unexploited until now. An efficient frequency upconverter for powerful microwave sources such as gyrotrons could find applications in tokamak plasma diagnostics, for example in collective Thomson scattering [2].

Theory:

As long as the frequency of the incident electromagnetic wave is of the same order or higher than the scattering frequency of the carriers, one can consider the simple equation of motion for single carriers, i.e. :

$$m^*(v)\dot{\vec{v}} + m^*(v)\vec{v}/\tau(v) = q\vec{E} \qquad (I)$$

$m^*(v)$: effective mass

$\tau(v)$: scattering time

A nonlinearity is introduced into this equation by the velocity dependence of the effective mass $m^*(v)$ (caused by the nonparabolicity of the conduction band) and the scattering time $\tau(v)$. The nonlinear motion of carriers of charge q with respect to the electric field E leads to a nonlinear polarization, which can be described by a power expansion [3] :

$$P = \chi E + \chi^{(2)}E^2 + \chi^{(3)}E^3 + ... \qquad (II)$$

χ : susceptibility

Since the free electron system is symmetric in space, $\chi^{(3)}$ is the first nonlinear coefficient differing from zero, leading to third harmonic generation.

The nonlinear polarization, $P(3\omega)$, is introduced into the Maxwellian equations as a source term, leading to [3] :

$$E(3\omega) = 4\pi\chi^{(3)}C(d)E^3(\omega) \qquad (III)$$

with (for perpendicular incidence)

$$C(d) = (e^{i3k(\omega)d} - e^{ik(3\omega)d}) / (\varepsilon(3\omega) - \varepsilon(\omega)) \qquad (IV)$$

$$k(\omega) = \sqrt{\varepsilon(\omega)}\ \omega/c$$

describing the dependence of the phase matching and absorption on the distance d from the surface. The linear optical properties, expressed by the complex dielectric function $\varepsilon(\omega)$, can be obtained using the Drude model:

$$\varepsilon(\omega) = \varepsilon_H + i4\pi\sigma(\omega)/\omega \qquad (V)$$

ε_H : background dielectric constant

$\sigma(\omega)$: conductivity

Since the conductivity is proportional to the carrier density, the absorption increases for higher doping levels of the material. This effect degrades the frequency tripling efficiency and compensates for the increase in $\chi^{(3)}$ due to higher doping [4].

Experimental Setup:

An optically-pumped gas laser was used to produce the high power far infrared radiation. The pump source is a two stage CO_2 laser system, which consists of a TEA-oscillator and a triple pass amplifier, operated at a pressure of 2 bars. The system is described in [2]. At the 9P20 line, used in these experiments, the laser produces μs pulses with energies up to 500J.

These pulses are coupled into the L-shaped FIR resonator passing through a wire grid, which acts as polarization-determining mirror for the far infrared. The output coupler is a flat mirror with a diameter of 200mm and a 80mm central hole. The FIR resonator is filled with CH_3F at a pressure of 3.5Torr, leading to an emission at a wavelength of 676µm, when pumped with a polarization parallel to the resonator polarization [5]. The maximum pulse energy for the FIR pulse was 120mJ, corresponding to a maximum power of 2MW. The laser is focussed onto the sample by a f=135mm Teflon lens, leading to a focal radius of about 2mm. The third harmonic radiation generated in the sample passes a waveguide cut-off filter [6], which consists of an array of 150µm holes, drilled into a 1mm thick metal plate. The transmission of the filter at the fundamental frequency is 10^{-10}, while it is about 0.6 at the third harmonic. For the detection of the third harmonic radiation we used either an InSb bolometer operated at 4.2K, which is very sensitive (pJ pulse energies can be detected) but slow, or a novel Ge hot-electron detector from the Ioffe institute, St. Petersburg. The Ge hot-electron detector operates at room temperature, has a temporal resolution of 1ns and a sensitivity of 1W. This detector allowed the time-resolved measurement of the third harmonic. The signal was recorded using a 2.5 Gsample/s digital oscilloscope. A second Ge hot-electron detector of the same type was used together with a beamsplitter to monitor the fundamental pulse. The sample was silicon, doped with $n=2.7*10^{15}/cm^3$. It was cut as a wedge, to prevent standing waves and to provide thickness-dependent measurements allowing a comparison with the theoretical C(d) function. The setup is shown in figure 1.

Results and discussion:

Fig. 2 shows a thickness-dependent measurement. The pulse energy of the third harmonic was measured time-integrated using the bolometer. While the pulse energy at the fundamental was kept constant, the crystal thickness was varied by translating the wedge cut sample. The measured points agree well with the theoretical $C^2(d)$ curve, which was calculated using measured absorption and calculated refractive index values and fitted in height. For the following experiments the optimum sample thickness of 0.5mm was used.

In Fig. 3 the results of a time-integrated measurement are shown, where the pulse energy at the fundamental frequency was progressively attenuated. Each point represents a single laser shot. Note that the pulse energy of the third harmonic could be observed over a range of more than 5 orders of magnitude, while the energy at the fundamental was varied over 2 orders of magnitude. For low pulse energies the dependence of the third harmonic signal from the fundamental shows a cubic power law as expected from the theoretical considerations. This behavior changes for pulse energies above 5mJ, corresponding to a peak power of about 100kW. The third harmonic then rises slower than the cube of the fundamental pulse energy. The time-integrated measurement averages over a range of powers, showing exceedingly large variations in particular at the third harmonic. Therefore a time-resolved experiment, where the powers are directly measured, should show saturation effects in a more pronounced way.

Fig. 4a, b shows the time resolved signals at 676μm and 225μm for a single laser shot. The temporal structure of the laser pulse shows peaks with a length of several ns, as expected. These peaks are due to beating of transversal modes. Axial mode beating would result in a beat frequency of 30 MHz which is not observed. The third harmonic peaks coincide and are

much more pronounced, thus clearly showing the nonlinear behavior. The relative peak heights, however, show some statistical fluctuations, compared to the peak heights at the fundamental. This is probably caused by spatial intensity fluctuations in the spot.

The heights of correlated peaks at the fundamental and third harmonic frequency for a number of shots are plotted against each other in Fig. 5. The highest peaks of the fundamental pulse reach a power of 2MW, which leads to 2kW of third harmonic power. Note that because of the poorer sensitivity of the fast detector, the power range shown here covers only the high power regime of the time-integrated experiment. In agreement with this we see a subcubic dependence of the third harmonic power on the fundamental power and, as expected, the saturation behavior is more pronounced with this direct measurement of power. The curve starts with an exponent of 2.5 at 100kW fundamental power and is reduced to 1.5 at the highest powers. The absorption of the sample, which was also measured, remained constant over the whole power range, showing that the noncubic intensity dependence of the third harmonic is caused by a diminution of $\chi^{(3)}$ at high power and not by nonlinear absorption. A possible explanation for an effect which reduces $\chi^{(3)}$ is that at high incident power the scattering rate of electrons is increased and exceeds the frequency of the incident radiation so that the former model of single electrons being accelerated by the electric field and performing a forced oscillation no longer holds. The nonlinear mechanism of harmonic generation is much more sensitive to a change in the transport properties than the linear absorption. For an incident power of more than 2MW, corresponding to an intensity of more than 15MW/cm^2, we observed an air breakdown in front of the sample, which inhibits a further rise of the conversion efficiency by an increase of fundamental power for this experimental setup.

Conclusions:

Time-resolved frequency tripling experiments with n-doped Si have been performed. For a power of 2MW at the fundamental frequency a conversion efficiency of 10^{-3} was reached. The dependence of the power at the third harmonic with respect to the power at the fundamental was subcubic for high power levels, showing saturation processes of the nonlinear effects. Breakdown limits in air at the sample surface were found for power densities above 15MW/cm^2.

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References:

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Figure Captions:

Fig.1:

Experimental setup; the elements of the FIR laser are: VV : vacuum vessel, PT : pyrex tube (20cm diameter), W1 : KCl window, W2 : TPX window, CM : concave aluminium coated glass mirror with RC=10m, WG : free standing wire grid, OC : flat metal output coupler with central hole of 80mm diameter, FM : focussing metal mirror, L : teflon lense with f=135mm, BS : beam splitter (mylar), S : wedged sample, F : waveguide cut-off filter, D1,D2 : identical Ge hot-electron detectors

Fig. 2:

pulse energy of the third harmonic vs. sample thickness; each dot represents a single laser shot. The curve shows the calculated $C^2(d)$ function, fitted in height.

Fig. 3:

pulse energy of the third harmonic vs. pulse energy of the fundamental, each dot representing a single laser shot

Fig. 4:

temporal evolution of the same laser shot at a) the fundamental frequency and b) the third harmonic

Fig. 5:

third harmonic power vs. fundamental power for correlated peaks (several shots)

Fig 1

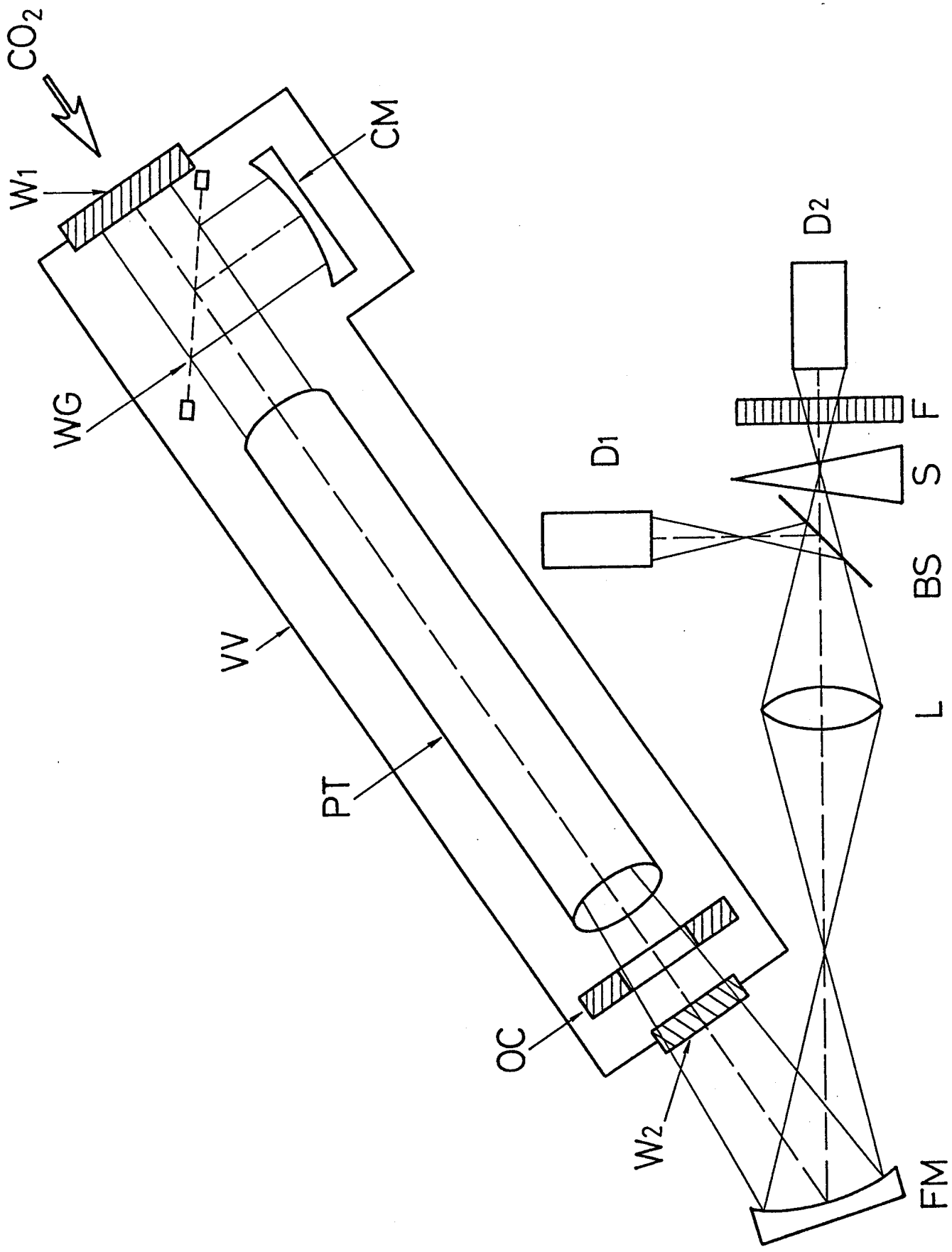

Fig 2

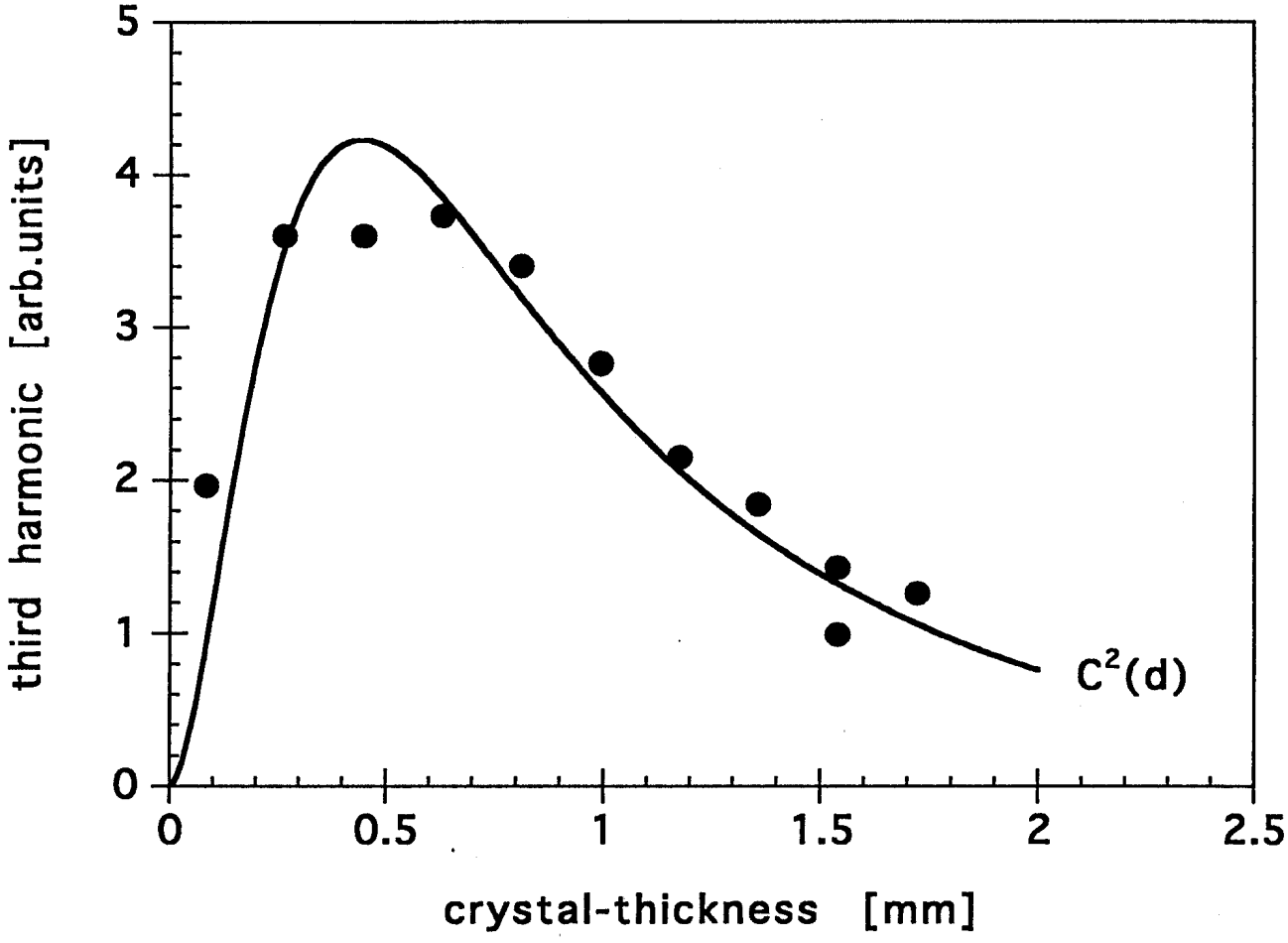
third harmonic [arb.units]
crystal-thickness [mm]
$C^2(d)$
0
1
2
3
4
5
0
0.5
1
1.5
2
2.5

Fig. 3

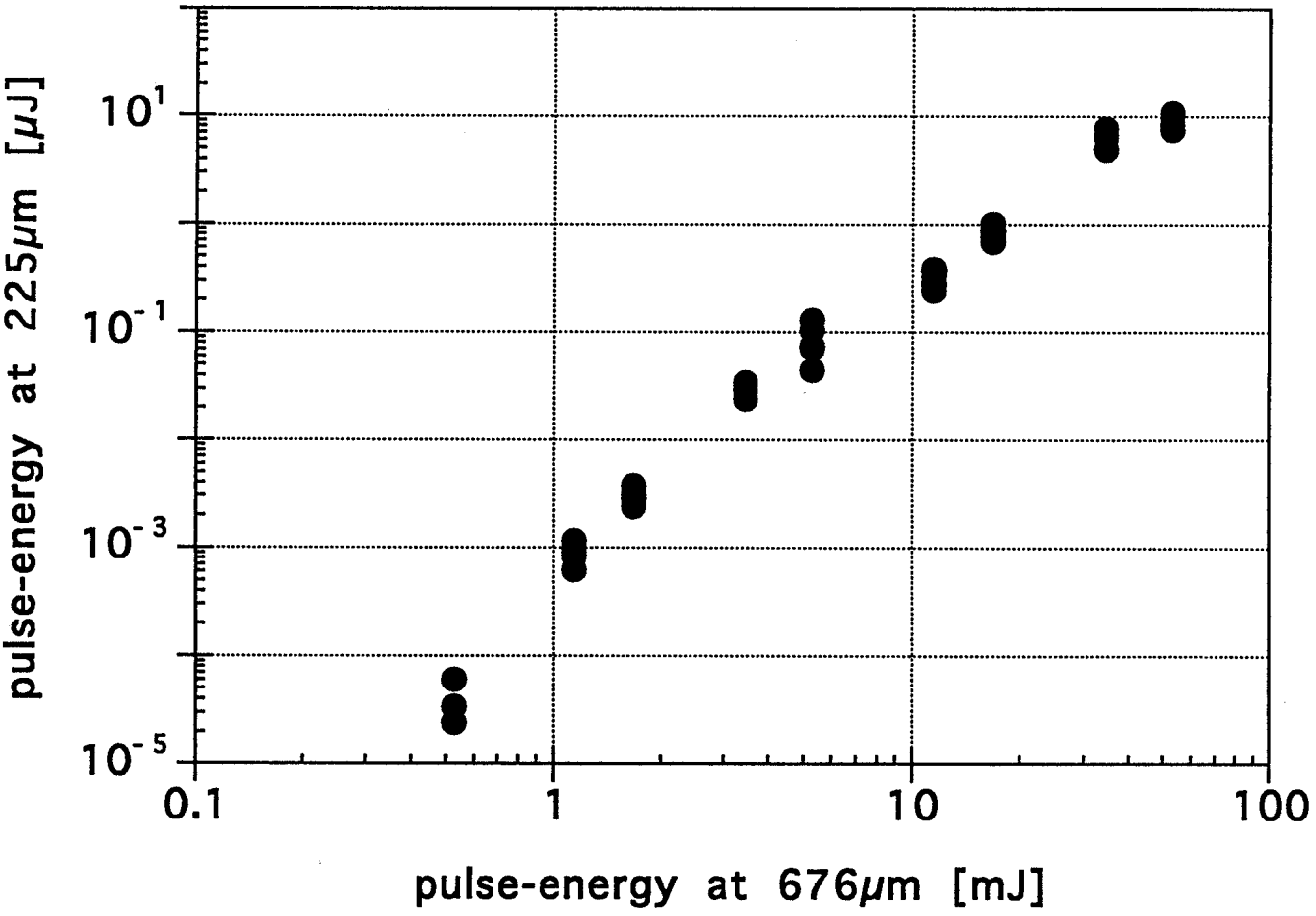
pulse-energy at 225μm [μJ]
10^1
10^-1
10^-3
10^-5
0.1
1
10
100
pulse-energy at 676μm [mJ]

Fig 4a

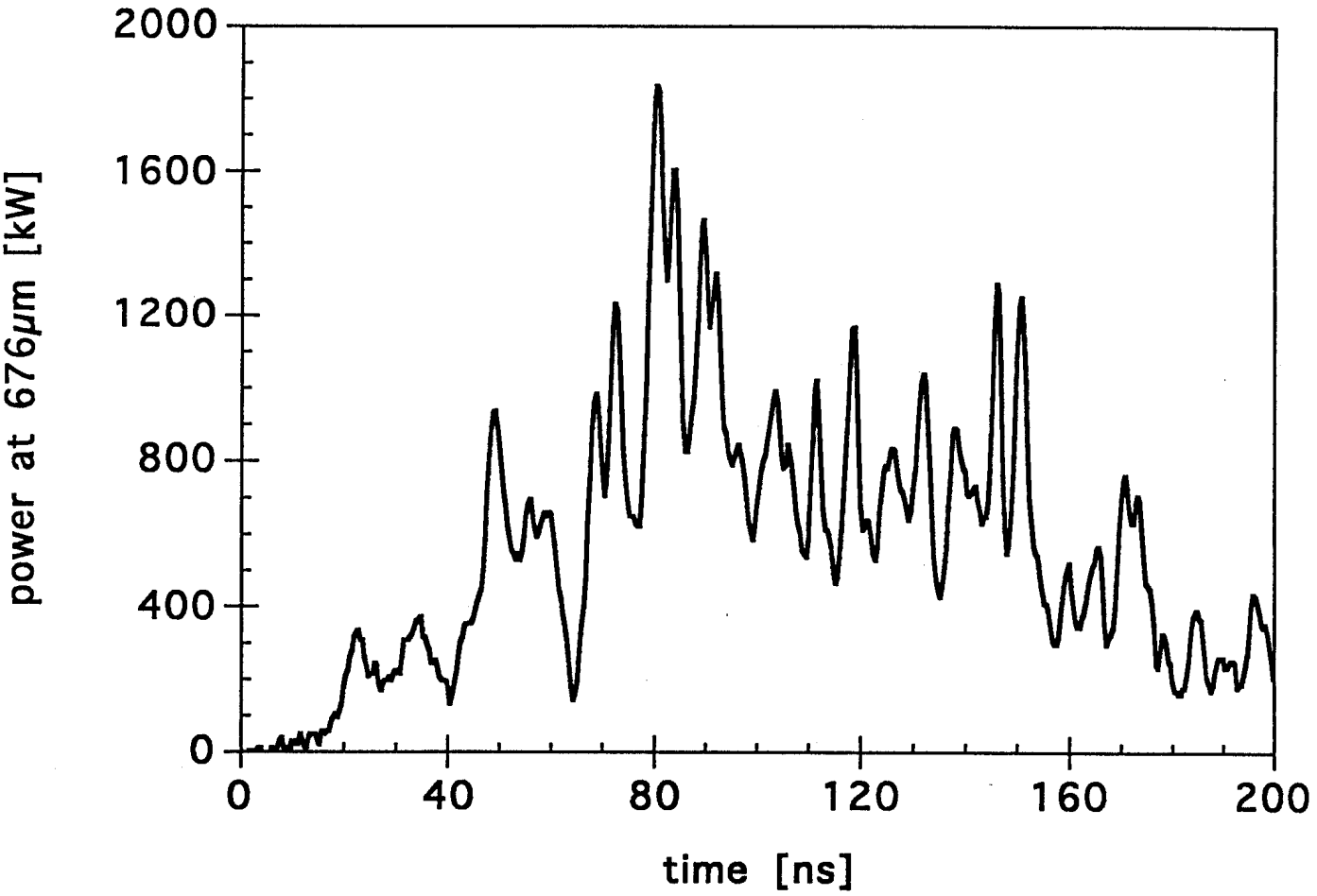
2000
1600
1200
800
400
0
power at 676µm [kW]
0
40
80
120
160
200
time [ns]

Fig 4b

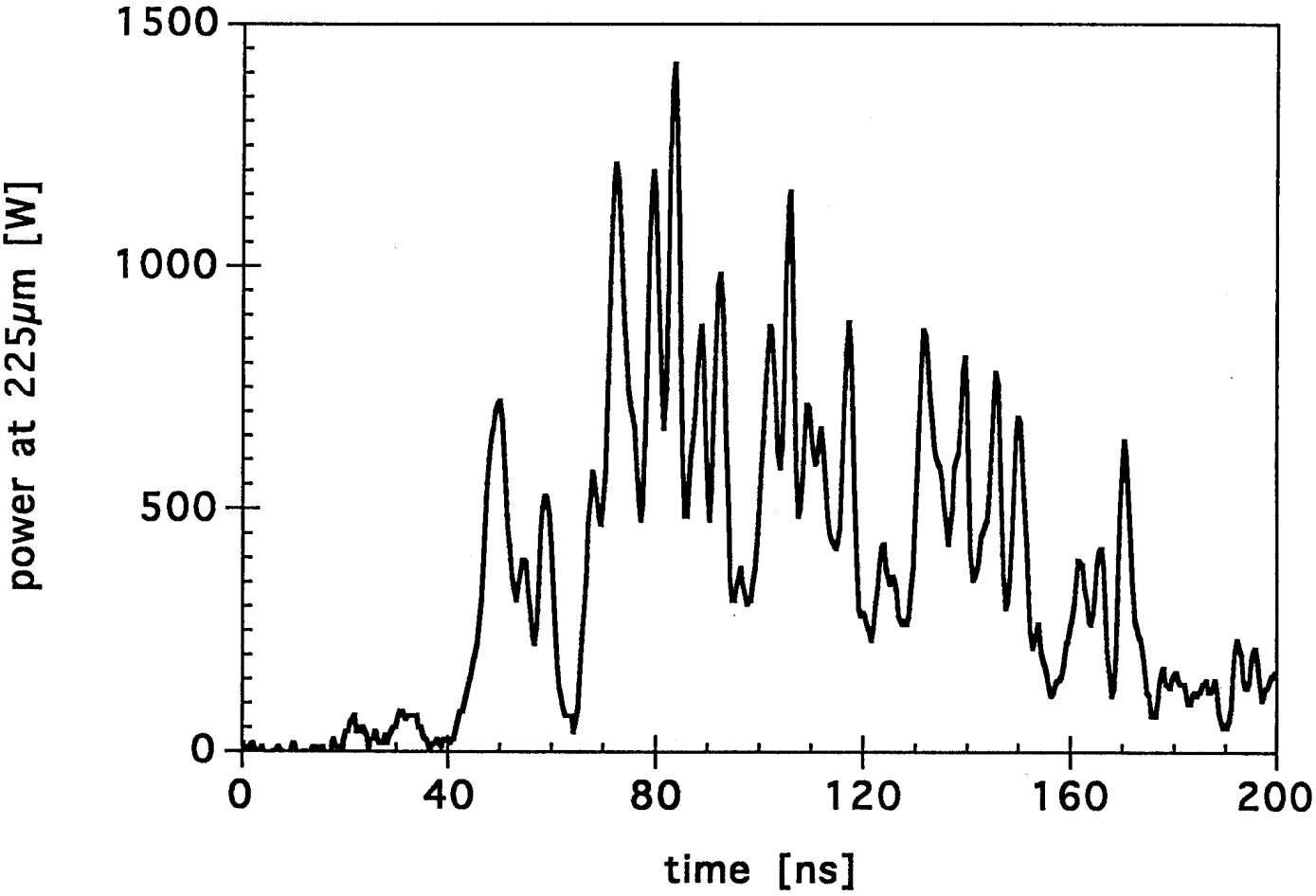

Fig 5